Evaluating Differences between Measured Personal Exposures to Volatile Organic Compounds and Concentrations in Outdoor and Indoor Air

KEN SEXTON,*,† JOHN L. ADGATE,‡ STEVEN J. MONGIN,‡ GREGORY C. PRATT,§ GURUMURTHY RAMACHANDRAN,‡ THOMAS H. STOCK,|| AND MARIA T. MORANDI||

School of Public Health, University of Texas, Brownsville Regional Campus, RAHC Building, 80 Fort Brown, Brownsville, Texas 78520, Division of Environmental & Occupational Health, School of Public Health, University of Minnesota, Minneapolis, Minnesota 55455, Minnesota Pollution Control Agency, 520 Lafayette Road, St. Paul, Minnesota 55155, and School of Public Health, University of Texas, Houston Health Science Center, Houston, Texas 77030

Accurate estimation of human exposures to volatile organic compounds (VOCs) is a key element of strategies designed to protect public health from the adverse effects of hazardous air pollutants. The focus here is on examining the capability of three different exposure metrics (outdoor community concentrations, indoor residential concentrations, and a simple time-weighted model) to estimate observed personal exposures to 14 VOCs. The analysis is based on 2-day average concentrations of individual VOCs measured concurrently in outdoor (O) air in three urban neighborhoods, indoor (I) air in participant's residences, and personal (P) air near the breathing zone of 71 healthy, nonsmoking adults. A median of four matched P–I–O samples was collected for each study participant in Minneapolis/St. Paul over three seasons (spring, summer, and fall) in 1999 using charcoal-based passive air samplers (3M model 3500 organic vapor monitors). Results show a clear pattern for the 14 VOCs, with P > I > O concentrations. Intra-individual variability typically spanned at least an order of magnitude, and inter-individual variability spanned 2 or more orders of magnitude for each of the 14 VOCs. Although both O and I concentrations generally underestimated personal exposures, I concentrations provided a substantially better estimate of measured P concentrations. Mean squared error (MSE) as well as correlation measures were used to assess estimator performance at the subject-specific level, and hierarchical, mixed effects models were used to estimate the bias and variance components of MSE by tertile of personal exposure. Bias and variance both tended to increase in the

* Corresponding author phone: (956)554-5165; fax: (956)554-5152; e-mail: ksexton@utb.edu.
† University of Texas, Brownsville.
‡ University of Minnesota.
§ Minnesota Pollution Control Agency.
|| University of Texas, Houston.

upper third of the P exposure distribution for O versus P and I versus P. A simple time-weighted model incorporating measured concentrations in both outdoor community air and indoor residential air provided no improvement over I concentration alone for the estimation of P exposure.

Introduction

Concentrations of hazardous air pollutants in a person's breathing zone for a defined period of time are typically referred to as *personal* exposures (*1*, *2*). An individual's personal exposure for a particular time period (e.g., 48 h) depends on pollutant concentrations in the indoor and outdoor microenvironments through which he or she moves during routine daily activities and on the time spent in each of these locations. From a public health perspective, it is often important to estimate the distribution of personal exposures in a population or to distinguish between individuals with high versus low exposure. But measuring personal exposures for a large number of people (including potentially vulnerable groups such as the young, the elderly, and the infirm) can be burdensome, time-consuming, expensive, and, in many cases, impractical. It is imperative, therefore, to gain a clear understanding of the value of more easily obtained metrics, such as measurements at outdoor community sites or indoor residential locations, for estimating personal exposures.

Although volatile organic compounds (VOCs) are common constituents of outdoor (*3–6*) and indoor (*7–9*) air, comparatively little is known about personal exposures. The relatively few personal monitoring studies that have been conducted suggest that personal exposures tend generally to be higher than indoor residential concentrations, which tend typically to be higher than outdoor community concentrations (*10–15*). Other personal exposure studies have concentrated on exposure of specific subpopulations to one or few individual VOCs, with many focused on exposure to benzene. This paper examines the ability of fixed indoor residential and outdoor monitors, in combination with time–activity data, to estimate personal exposures to 14 individual VOCs for 71 nonsmoking adults in three urban neighborhoods.

Study Design

The study was designed primarily to measure exposures to VOCs experienced by healthy, nonsmoking adults and to compare results with concurrent measurements inside their residences and outside in their neighborhoods (*13*). A secondary objective was to measure $PM_{2.5}$ exposures for a subset of the subjects, results of which have been published previously (*16–19*). Three urban neighborhoods (Phillips, East St. Paul, and Battle Creek) with different outdoor VOC concentration profiles based on modeling results (*13*, *20*, *21*) were selected for the exposure monitoring study.

Phillips (PHI) is an economically disadvantaged, predominantly minority inner-city neighborhood in south central Minneapolis. Outdoor VOC concentrations in PHI were predicted to be relatively high because of contributions from multiple sources. East St. Paul (ESP) is a blue-collar, racially mixed neighborhood in St. Paul. VOC concentrations were predicted to be relatively high, primarily as a result of emissions from nearby manufacturing plants. Battle Creek (BCK) is a predominantly white, affluent neighborhood on the eastern edge of St. Paul. Predicted VOC concentrations were relatively low as compared to the other two neighborhoods.

A centralized outdoor monitoring site (community site) was established in each neighborhood. Approximately 25 healthy, nonsmoking adults were recruited from each neighborhood using house-to-house canvassing and direct solicitation (informed consent was obtained). Matched 2-day samples were collected outdoors at the three community monitoring sites, indoors in participants' residences (room where they spent most of their waking hours), and near participants' breathing zones, all using passive dosimeters. Participants also completed time−activity logs recording the time they spent in seven microenvironments (indoors at home, work, other; outdoors at home, work, other; in transit) and the time they were in close proximity to environmental tobacco smoke. All outdoor community site (O), indoor residential (I), and personal (P) samples were collected during three monitoring sessions in 1999: spring (April 26−June 20); summer (June 21−August 11); and fall (September 23−November 21).

Methods

VOC Collection and Analysis. All VOC concentrations (O, I, and P) were 2-day (approximately 48-h) average values obtained with 3M model 3500 organic vapor monitors (3500 OVMs), which are charcoal-based passive air samplers. The suitability of these VOC badges for outdoor, indoor, and personal sampling has been demonstrated by Chung et al. (*22, 23*). These investigators have also described the determination of extraction efficiencies and the calculation of method detection limits. The extraction solvent consisted of a 2:1 v/v mix of acetone and carbon disulfide, which provided a very low background for target analytes. All extracts were analyzed by GC/MS with a Hewlett-Packard 5890 series II Plus GC with an HP 5972 MS detector, HP 18593B autosampler, and Vectra 486 computer with EnvironQuant Chem-Station Software and NBS75K Spectra Library, using an RTX-1/60 m/0.25 mm i.d./1 mm film thickness capillary column. Analytical and internal standards were prepared, and VOC concentrations were calculated as described previously (*23*). Duplicate O, I, and P badges were collected periodically during the study (total $n = 80$), and correlation coefficients for the positive measurements were >0.95 for all individual VOCs except styrene (0.94) and chloroform (0.95). We define the median relative absolute difference (MRAD) as the median of the ratios of within-pair absolute differences divided by the within-pair mean. MRAD was <0.18 for all VOCs except trichloroethylene (0.44).

Statistical Analyses. All statistical analyses were performed using SAS (*24*) and S-plus (*25*). Concentrations less than the method detection limit (MDL) were included in the calculations. "Nondetectable" measurements (i.e., samples with no analytical response or those with values ≤0 after blank subtraction) were assigned a value of one-half the analytical detection limit (ADL).

Three estimators of personal exposure are evaluated:

$$\hat{P}_{[O]ij} = O_{ij} \tag{1}$$

$$\hat{P}_{[I]ij} = I_{ij} \tag{2}$$

$$\hat{P}_{[OI]ij} = O_{ij}t_{[O]ij} + I_{ij}t_{[I]ij} \tag{3}$$

where O_{ij} and I_{ij} denote the observed concentration for the *i*th subject on the *j*th occasion, from O and I, respectively; $t_{[O]ij}$ and $t_{[I]ij}$ represent the (time) fraction of the 2-day monitoring period spent in the O and I environments, respectively. For the purposes of this analysis, we let $\hat{P}_{[O]}$, $\hat{P}_{[I]}$, and $\hat{P}_{[OI]}$ represent $\hat{P}_{[O]ij}$, $\hat{P}_{[I]ij}$, and $\hat{P}_{[OI]ij}$ for all subjects and all times. In the simple time-weighted model (eq 3), the proportion of time in O was defined to be the complement of the proportion of time in I (i.e., $t_{[O]ij} = 1 - t_{[I]ij}$). Thus, the

model implicitly assumes that individuals not in their homes are exposed uniformly to the measured O concentration regardless of whether they are indoors or out (a likely underestimate of actual exposures).

The mean squared error (MSE) was computed for each estimator of P (see Discussion). To maintain the original scale of measurement, the root mean squared error (RMSE) is reported instead of the MSE, and the variance is represented by its square root as the standard deviation (SD). For $\hat{P}_{[O]}$ and $\hat{P}_{[I]}$, both RMSE and longitudinal correlations are first presented, based on estimation of each statistic for each subject. Geometric means of all duplicate samples are used to facilitate comparison with previous studies. To maximize the information available from our sample, no lower limit on the number of repeated measurements was applied in the calculation of these subject-specific RMSEs or longitudinal correlations (*R*) except for the mathematical limit imposed by the statistics themselves. For example, a longitudinal correlation cannot be calculated for subjects with only one data point. We report medians with upper and lower deciles to illustrate the inter-subject distributions of RMSE and *R* and to give a sense of their inherent variation.

To further analyze the MSE by its component bias and variance, mixed effects models were used (*26, 27*). These models accommodate duplicate data explicitly, making use of information on measurement error, and obviating the need to take means of duplicates. Moreover, mixed effects models handle variation in numbers of measurements across subjects by downweighting those with fewer measurements. To allow for heterogeneity of variance and to more flexibly model bias, the range of all P exposures was divided into its three tertiles, and a separate model was fitted for each.

Results

Selected sociodemographic characteristics and exposure-related attributes for the 71 participants in the study are summarized in Table 1. Seventy-seven percent were female, and more than half (56%) were between the ages of 40−65 years. Only one person had less than a high school education, while 37% had some college, 18% were college graduates, and 34% reported some post-graduate education. More than half (51%) had an annual household income of $40 000 or more, with 8% earning between $75 000 and $100 000, and 3% earning more than $100 000. Eighty-five percent were white, 7% were African American, 3% were Native American, 1% was Asian/Pacific Islander, and 3% were other. Most participants (69%) worked outside the home, only 7% lived with a smoker, and 34% had attached garages. Overall, the participants were predominantly white, female, well-educated, relatively affluent, and unlikely to be exposed to environmental tobacco smoke at home. As expected, participants from PHI had the highest percentage of minorities and the lowest household incomes.

A total of 284 valid, matched P and O sample sets with time−activity diaries were obtained from the 71 participants. The number collected for each participant varied from 1 (2 people) to 11 (1 person), with a median of 4 (Q25 = 3 and Q75 = 6). This variability in the number of P samples for each individual results from the fact that some participants dropped out of the study early, while others continued to participate. Furthermore, some of the P samples were invalid because of protocol errors, monitor malfunctions, or analytical problems. The range of P VOC concentrations for each participant is displayed graphically in Figure 1, and the range of I VOC concentrations for each participant is presented in Figure 2. For individual VOCs, each line in these range plots represents one of the 71 participants and spans the range of concentrations measured for that person. Within each VOC, the line segments are ordered by maximum concentration, and the vertical ordering of the VOCs themselves is deter-

TABLE 1. Summary of Sociodemographic Information for Participants in the Study

		n (%)		
parameter	Battle Creek	East St. Paul	Phillips	all communities
no. of participants	25 (35%)	22 (31%)	24 (34%)	71 (100%)
gender				
male	7 (10%)	2 (3%)	7 (10%)	16 (23%)
female	18 (25%)	20 (28%)	17 (24%)	55 (77%)
age				
18−39 yr	6 (8%)	12 (17%)	11 (15%)	29 (41%)
40−65 yr	17 (24%)	10 (14%)	13 (18%)	40 (56%)
>65 yr	1 (1%)	0 (0%)	0 (0%)	1 (1%)
missing	1 (1%)	0 (0%)	0 (0%)	1 (1%)
education				
less than high school	0 (0%)	1 (1%)	0 (0%)	1 (1%)
high school	2 (3%)	2 (3%)	3 (4%)	7 (10%)
some college	7 (10%)	13 (18%)	6 (8%)	26 (37%)
college graduate	6 (8%)	2 (3%)	5 (7%)	13 (18%)
post-graduate education	10 (14%)	4 (6%)	10 (14%)	24 (34%)
annual household income				
$10 000−$19 999	2 (3%)	1 (1%)	3 (4%)	6 (8%)
$20 000−$29 999	0 (0%)	4 (6%)	9 (13%)	13 (18%)
$30 000−$39 999	4 (6%)	3 (4%)	3 (4%)	10 (14%)
$40 000−$49 999	2 (3%)	6 (8%)	1 (1%)	9 (13%)
$50 000−$74 999	9 (13%)	5 (7%)	5 (7%)	19 (27%)
$75 000−$99 999	2 (3%)	3 (4%)	1 (1%)	6 (8%)
≥$100 000	2 (3%)	0 (0%)	0 (0%)	2 (3%)
refused or missing	4 (6%)	0 (0%)	2 (3%)	6 (8%)
race				
white	24 (34%)	22 (31%)	14 (20%)	60 (85%)
African American	1 (1%)	0 (0%)	4 (6%)	5 (7%)
Native American	0 (0%)	0 (0%)	2 (3%)	2 (3%)
Asian/Pacific Islander	0 (0%)	0 (0%)	1 (1%)	1 (1%)
other	0 (0%)	0 (0%)	2 (3%)	2 (3%)
refused	0 (0%)	0 (0%)	1 (1%)	1 (1%)
work outside the home				
yes	17 (24%)	13 (18%)	19 (27%)	49 (69%)
no	8 (11%)	9 (13%)	5 (7%)	22 (31%)
live with a smoker				
yes	1 (1%)	3 (4%)	1 (1%)	5 (7%)
no	24 (34%)	19 (27%)	23 (32%)	66 (93%)
attached garage				
yes	18 (25%)	3 (4%)	3 (4%)	24 (34%)
no	7 (10%)	19 (27%)	21 (30%)	47 (66%)

mined by the median of these maximum values. Minimum values appear to be truncated in some cases (for example, β-pinene, chloroform, and styrene in Figure 1) because many samples were zero or below.

As shown in Figure 1, a participant's P exposure to individual VOCs over multiple monitoring periods (within-person variability) often spanned 1 or more orders of magnitude. Moreover, the difference between participants' with the lowest maximum P values and those with the highest (between-person variability) often spanned 2 or more orders of magnitude. This same pattern also held true for participants' I concentrations (Figure 2), which were generally lower than matched P exposures. The evidence indicates that for these 14 VOCs there was substantial within-person variability and between-person variability for both P exposures and I concentrations.

Two subject-level criteria for characterizing the performance of $\hat{P}_{[O]}$ are provided in Table 2. The RMSE and R were calculated for each subject with a sufficient number of samples for each measure (≥ 1 for RMSE, ≥ 2 for R). RMSE is a measure of the magnitude and variation of the difference ($\mu g/m^3$) between measured O concentrations and P exposures, while R is a measure of the linear association between O and P. Compared to R and for the aims of this study, RMSE is a more direct measure of performance of these estimators (see Discussion); however, we present both to facilitate their comparison. The median value for RMSE and R across all subjects along with 10th and 90th percentiles are presented.

Looking at the overall results, the median RMSE for $\hat{P}_{[O]}$ was between 0.2 and 1.8 $\mu g/m^3$ for 9 VOCs, between 2.5 and 4.8 $\mu g/m^3$ for 3 VOCs (α-pinene, β-pinene, and m-/p-xylene), 13.4 $\mu g/m^3$ for D-limonene, and 16.3 $\mu g/m^3$ for toluene. It should be noted that RMSE is expected to be elevated for those VOCs found at higher concentrations, since their variance is usually higher as well. For all 14 VOCs, $\hat{P}_{[O]}$ underestimated P exposure. The RMSE of $\hat{P}_{[O]}$ for 6 VOCs (benzene, ethyl benzene, methylene chloride, toluene, o-xylene, m-/p-xylene) was substantially higher in BCK than in ESP and PHI. This is not surprising given that O concentrations in BCK tended to be slightly but consistently lower than in the other two neighborhoods, while P exposures tended to be slightly higher (hence a greater underestimation of P exposures in BCK). Overall, correlation coefficients were generally unremarkable, with median R for 9 VOCs between −0.08 and 0.24 and between 0.43 and 0.59 for the other 5 VOCs (benzene, ethyl benzene, styrene, o-xylene, m-/p-xylene). With the exceptions of D-limonene and tetrachloroethylene in ESP and benzene, styrene, and m-/p-xylene in BCK, R values were generally comparable across the three neighborhoods.

Analogous performance measures for $\hat{P}_{[I]}$ by neighborhood are provided in Table 3. Comparing $\hat{P}_{[I]}$ to $\hat{P}_{[O]}$ overall, there was a reduction in RMSE for 13 of the 14 VOCs (trichloroethylene remained unchanged). The most dramatic reductions were observed for D-limonene (from 13.4 to 4.7 $\mu g/m^3$) and toluene (from 16.3 to 8.3 $\mu g/m^3$). Generally, $\hat{P}_{[I]}$ also
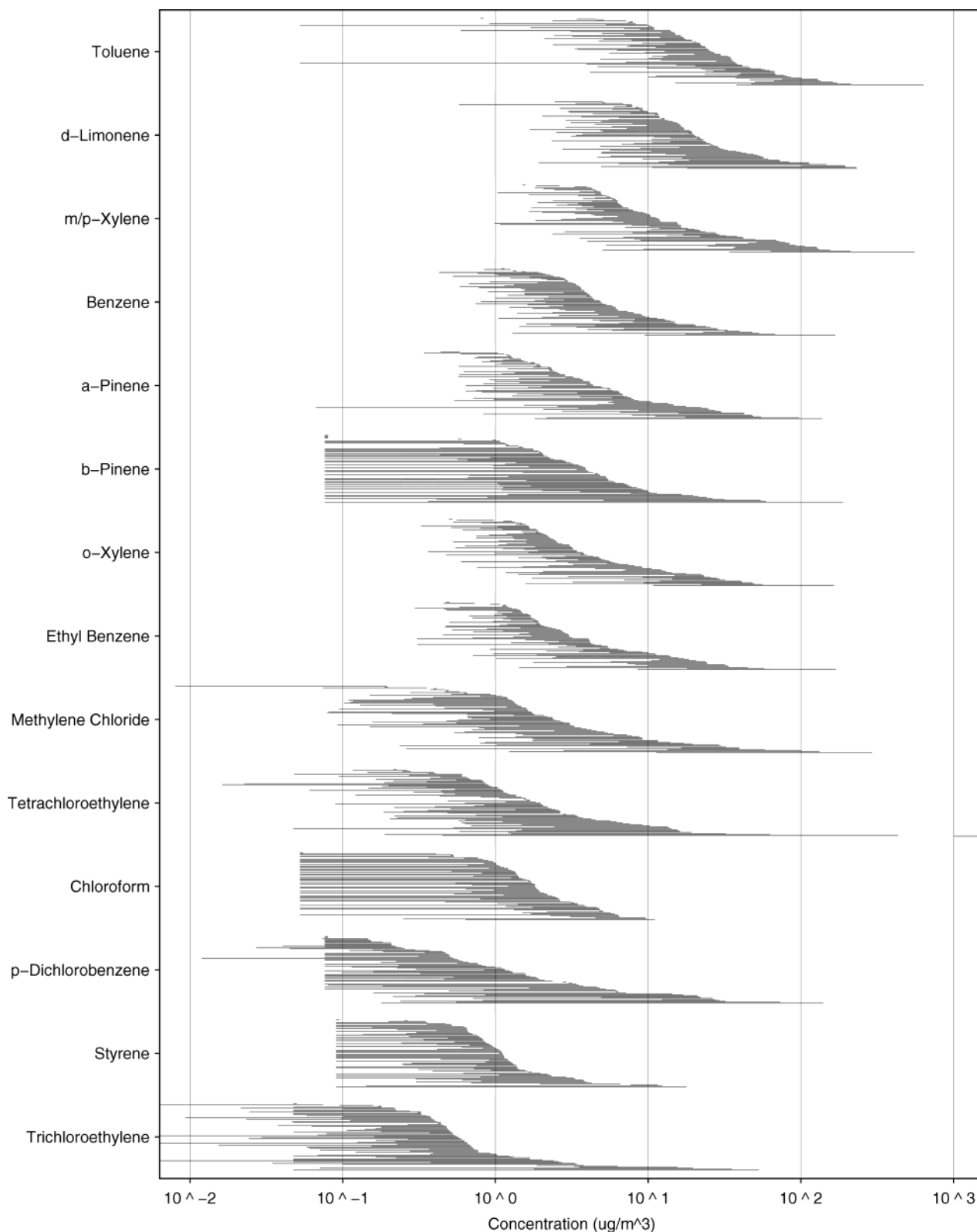
FIGURE 1. Range plot of measured personal VOC exposures for each study participant.

tended to underestimate P exposures, but not as much or as consistently as $\hat{P}_{[O]}$. Compared to $\hat{P}_{[O]}$, reduced RMSE occurred similarly across neighborhoods, and for benzene, toluene, *o*-xylene, and *m*-/*p*-xylene remained higher in BCK as compared to ESP and PHI (similar to the pattern observed for $\hat{P}_{[O]}$). Correlation coefficients improved dramatically for all VOCs, with $R > 0.85$ for 8 VOCs (benzene, chloroform, D-limonene, methylene chloride, α-pinene,

β-pinene, *o*-xylene, *m*-/*p*-xylene), and $0.57 \leq R \leq 0.83$ for the remaining 6 VOCs. Relatively consistent R values for individual VOCs were observed across neighborhoods, with the exception of *p*-dichlorobenzene (0.16) in BCK and trichloroethylene (0.40) in PHI.

The improved performance of I over O concentrations is illustrated graphically in Figure 3 using three VOCs as examples. It is clear from the scatter plots that O concentra-
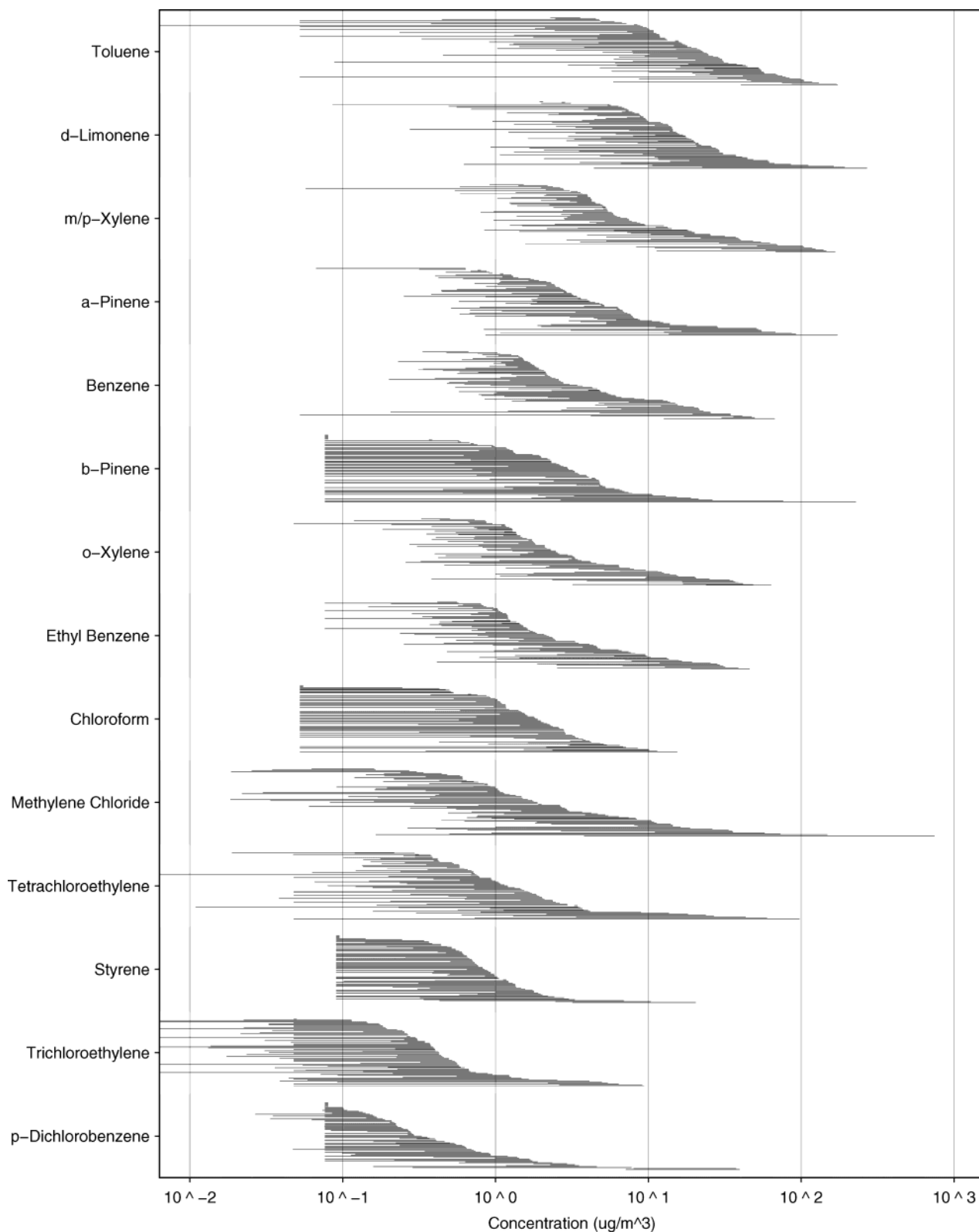
FIGURE 2. Range plot of measured indoor residential VOC concentrations for each study participant.

tions underestimate P exposure in the majority of cases for benzene, methylene chloride, and toluene. I concentrations, on the other hand, provide a noticeable improvement in estimating P exposure, although the tendency is still to underestimate. Overall, for this population and under the conditions of the study, matched I concentrations provided a substantially better estimate of personal VOC exposure than matched O concentrations for all 14 VOCs measured.

To further investigate the nature of RMSE for $\hat{P}_{[O]}$ and $\hat{P}_{[I]}$, we divided the domains of measured P exposures into lower, middle, and upper tertiles and then estimated the two components of MSE, bias (squared) and variance, using the mixed effects model. We distinguish the vector of all numerical observations of personal exposures by **P**. An estimate of bias is obtained for each tertile by estimating the mean of $\hat{P}_{[O]} - \mathbf{P}$ (Table 4) or the mean of $\hat{P}_{[I]} - \mathbf{P}$ (Table 5).

TABLE 2. Comparison of Matched (Outdoor Community, Personal) VOC Concentrations for Individual Participants in the Study

compound	Battle Creek		East St. Paul		Phillips		all communities	
	RMSE[a]	R[b]	RMSE	R	RMSE	R	RMSE	R
benzene	4.9	0.02	1.4	0.79	1.5	0.68	1.8	0.59
	(0.9, 19.4)[c]	(−0.92, 0.89)[c]	(0.9, 7.8)	(−0.37, 1.00)	(0.7, 8.5)	(−0.92, 1.00)	(0.7, 16.3)	(−0.85, 1.00)
chloroform	1.1	0.00	1.4	0.31	1.5	0.00	1.8	0.00
	(0.3, 3.3)	(−0.51, 1.00)	(0.6, 4.6)	(−0.16, 0.92)	(0.5, 4.2)	(−0.54, 0.98)	(0.5, 3.9)	(−0.50, 0.99)
p-dichlorobenzene	0.6	0.00	0.3	0.15	0.9	−0.01	0.7	0.00
	(0.1, 9.5)	(−0.54, 0.58)	(0.1, 8.4)	(−0.73, 0.82)	(0.1, 13.5)	(−0.78, 1.00)	(0.1, 9.8)	(−0.72, 0.98)
ethyl benzene	3.7	0.41	1.1	0.51	1.1	0.65	1.5	0.53
	(1.0, 14.3)	(−0.47, 1.00)	(0.5, 13.4)	(−0.76, 1.00)	(0.6, 12.3)	(0.01, 1.00)	(0.5, 14.3)	(−0.52, 1.00)
D-limonene	12.7	0.43	17.5	−0.31	12.2	0.34	13.4	0.15
	(6.3, 36.2)	(−0.64, 1.00)	(7.6, 86.5)	(−0.97, 1.00)	(6.9, 40.6)	(−1.00, 1.00)	(7.0, 57.2)	(−1.00, 1.00)
methylene chloride	3.3	0.14	1.5	0.11	0.8	0.38	1.3	0.14
	(0.2, 32.0)	(−0.93, 0.96)	(0.4, 8.3)	(−0.90, 0.84)	(0.4, 8.3)	(−0.76, 1.00)	(0.3, 12.4)	(−0.86, 1.00)
α-pinene	3.6	−0.04	3.6	0.12	2.0	−0.17	3.0	−0.08
	(1.2, 17.9)	(−0.93, 0.95)	(1.1, 15.4)	(−0.87, 0.96)	(1.2, 14.6)	(−1.00, 0.75)	(1.2, 17.3)	(−1.00, 0.95)
β-pinene	1.8	0.00	3.6	0.00	1.9	0.28	2.5	0.00
	(0.1, 8.8)	(−0.13, 0.98)	(1.5, 9.5)	(−0.40, 0.87)	(0.8, 7.2)	(0.00, 1.00)	(0.8, 9.4)	(−0.16, 0.99)
styrene	0.6	0.08	0.5	0.74	0.6	0.40	0.6	0.55
	(0.2, 2.1)	(−0.20, 0.99)	(0.2, 2.3)	(0.20, 0.98)	(0.4, 1.7)	(−0.82, 0.90)	(0.2, 2.0)	(−0.23, 0.98)
tetrachloroethylene	1.3	−0.15	1.0	0.62	0.6	0.17	0.9	0.24
	(0.2, 25.3)	(−0.75, 1.00)	(0.1, 8.7)	(−0.64, 0.99)	(0.2, 3.7)	(−1.00, 0.98)	(0.2, 8.9)	(−0.91, 1.00)
toluene	29.9	−0.11	13.8	0.06	10.0	0.33	16.3	0.02
	(9.6, 85.2)	(−0.99, 1.00)	(7.4, 50.4)	(−0.65, 0.87)	(5.1, 19.3)	(−0.49, 1.00)	(6.1, 64.5)	(−0.90, 0.99)
trichloroethylene	0.2	0.16	0.2	0.43	0.2	0.20	0.2	0.24
	(0.1, 1.1)	(−0.65, 1.00)	(0.1, 1.5)	(−0.16, 0.98)	(0.1, 0.5)	(−0.92, 0.98)	(0.1, 1.3)	(−0.79, 0.99)
o-xylene	4.2	0.26	1.3	0.44	1.3	0.67	1.6	0.43
	(1.2, 17.3)	(−0.94, 0.96)	(0.5, 16.5)	(−0.78, 0.97)	(0.6, 16.3)	(0.13, 1.00)	(0.6, 19.0)	(−0.72, 1.00)
m-/p-xylene	14.1	0.19	3.8	0.50	3.7	0.70	4.8	0.47
	(3.4, 57.4)	(−0.75, 0.98)	(1.5, 51.9)	(−0.76, 0.99)	(1.8, 54.0)	(0.08, 1.00)	(2.0, 63.4)	(−0.69, 1.00)

[a] Median root mean squared error ($\mu g/m^3$); n for number of research subjects = 25 for Battle Creek, 22 for East St. Paul, and 24 for Phillips. [b] Median correlation coefficient; n for number of research subjects varies from 20 to 23 for Battle Creek, n = 22 for East St. Paul, and n varies from 21 to 22 for Phillips. [c] 10th and 90th percentiles.

In addition, an estimate of standard deviation (SD) is obtained from the square root of the variance resulting from application of the same mixed model.

The results from Table 4 show that both the bias and the SD of $\hat{P}_{[O]}$ tend to increase from the lower to the middle and from the middle to the upper tertile of the distribution of P exposures. For example, in the lower tertile the range of $\hat{P}_{[O]}$ − **P** values for individual VOCs is between −1.9 and 0.0; in the middle tertile it is between −13.2 and −0.1; and in the upper tertile it is between −62.7 and −2.0. These data suggest that $\hat{P}_{[O]}$ typically underestimates P exposures in all cases and that the magnitude of this underestimation increases with higher P exposures. In terms of variance, the range of SDs for individual VOCs in the lower tertile is 0.1−9.9, 0.2−6.4 in the middle tertile, and 1.9−360 in the upper-tertile. Again, the data indicate that the variance of $\hat{P}_{[O]}$ generally increases with higher P exposures.

The pattern is similar for I versus P in Table 5, with both bias and variance of $\hat{P}_{[I]}$ increasing from lower to middle to higher tertiles of P exposures. For example, the range of $\hat{P}_{[I]}$ − **P** values for the 14 individual VOCs is −0.9 to 0.4 in the lower tertile, −3.3 to 1.1 in the middle tertile, and −55.8 to 3.9 in the upper tertile. The evidence suggests that $\hat{P}_{[I]}$ tends to underestimate P exposures for 12 of the 14 VOCs, especially in upper tertile of the distribution of P exposures. $\hat{P}_{[I]}$ tends to overestimate, particularly in the upper tertile, for methylene chloride and α-pinene. Similarly, the range of SDs for individual VOCs increases from 0.1 to 5.2 in the lower tertile, from 0.2 to 10.2 in the middle tertile, and from 1.7 to 351 in the upper tertile, which suggests that variance in $\hat{P}_{[I]}$ also tends to increase with higher P exposures.

We investigated the time-weighted estimator $\hat{P}_{[OI]}$ by examining the RMSE for the model where P exposure equals the time fraction of the 2-day monitoring period spent indoors at home ($t_{[I]}$) times the measured I concentration plus the complement of the time fraction indoors at home ($1 − t_{[I]}$) times the measured O concentration (see eq 3). Because the measured O concentration is likely to be less than or equal to unmeasured concentrations in the other microenvironments, it represents quasi-baseline conditions (i.e., minimal exposures) when participants were not inside their homes. In Table 6, the RMSE for this model is apportioned into bias and SD using the same approach as for $\hat{P}_{[O]}$ and $\hat{P}_{[I]}$ (Tables 4 and 5).

As found for $\hat{P}_{[O]}$ and $\hat{P}_{[I]}$, the bias (the expected difference between the estimator and P exposure) to noise (SD) ratio for $\hat{P}_{[OI]}$ in Table 6 is relatively low. Nevertheless, it is apparent that the estimated bias of $\hat{P}_{[OI]}$ was similar to that for $\hat{P}_{[I]}$ (Table 5) in the lower and middle tertiles. At the higher tertile, the bias was greater for the time-weighted estimator in every instance except methylene chloride. The SD was generally similar between $\hat{P}_{[I]}$ and the time-weighted model across all three tertiles.

Discussion

Chronic exposure to relatively low levels of airborne VOCs is an inescapable reality for residents of the United States. This class of chemicals is ubiquitous in occupational and nonoccupational settings, including both indoor and outdoor environments. Not only are VOCs released into the air from industrial processes, internal combustion engines, cigarette smoking, and bathing or showering in chlorinated water, they are also common constituents in cleaning and degreasing agents, deordorizers, dry-cleaning processes, paints, pesticides, personal care products, and solvents (2, 7−15). Of the 14 VOCs measured in this study, five originate from primarily indoor sources (chloroform, p-dichlorobenzene, D-limonene, and α- and β-pinene), while nine are emitted by a combination of indoor and outdoor sources (benzene,

TABLE 3. Comparison of Matched (Indoor Residential, Personal) VOC Concentrations for Individual Participants in the Study

	Battle Creek		East St. Paul		Phillips		all communities	
compound	RMSE[a]	R[b]	RMSE	R	RMSE	R	RMSE	R
benzene	2.5	0.86	1.6	0.89	1.1	0.78	1.7	0.86
	(0.5, 9.7)[c]	(0.12, 1.00)[c]	(0.4, 6.0)	(0.10, 1.00)	(0.6, 4.4)	(−0.98, 1.00)	(0.4, 8.1)	(−0.26, 1.00)
chloroform	0.4	0.89	0.6	0.90	0.5	0.70	0.5	0.88
	(0.2, 1.5)	(0.06, 1.00)	(0.3, 1.7)	(−0.19, 0.99)	(0.3, 2.7)	(−0.32, 1.00)	(0.2, 1.7)	(−0.05, 1.00)
p-dichlorobenzene	0.5	0.16	0.2	0.64	0.5	0.62	0.3	0.57
	(0.1, 9.0)	(−0.42, 0.99)	(0.1, 8.3)	(−0.48, 0.99)	(0.1, 6.8)	(−0.88, 1.00)	(0.0, 9.0)	(−0.54, 1.00)
ethyl benzene	1.4	0.69	0.8	0.85	0.8	0.73	1.0	0.75
	(0.3, 11.0)	(−0.94, 0.99)	(0.2, 3.0)	(−0.26, 1.00)	(0.3, 15.6)	(−0.13, 1.00)	(0.3, 11.1)	(−0.39, 1.00)
D-limonene	4.2	0.96	5.8	0.98	4.7	0.94	4.7	0.96
	(2.1, 18.9)	(0.34, 1.00)	(2.5, 36.3)	(0.26, 1.00)	(1.8, 45.0)	(−0.84, 1.00)	(2.1, 36.4)	(0.11, 1.00)
methylene chloride	1.3	0.95	0.6	0.93	0.8	0.81	0.8	0.90
	(0.3, 26.2)	(−0.37, 1.00)	(0.2, 1.8)	(0.37, 1.00)	(0.2, 5.7)	(−0.09, 1.00)	(0.2, 8.7)	(−0.04, 1.00)
α-pinene	1.3	0.98	0.8	0.92	1.0	0.92	1.0	0.95
	(0.3, 7.2)	(0.34, 1.00)	(0.3, 6.3)	(0.61, 1.00)	(0.2, 14.0)	(−0.51, 1.00)	(0.2, 7.6)	(−0.42, 1.00)
β-pinene	0.9	0.98	1.2	0.96	1.1	0.97	1.0	0.97
	(0.1, 2.8)	(0.00, 1.00)	(0.3, 4.9)	(0.15, 1.00)	(0.4, 2.9)	(0.00, 1.00)	(0.2, 4.1)	(0.00, 1.00)
styrene	0.4	0.65	0.3	0.70	0.4	0.77	0.4	0.71
	(0.1, 1.2)	(−0.19, 1.00)	(0.2, 1.3)	(0.01, 0.99)	(0.2, 1.1)	(0.01, 1.00)	(0.2, 1.2)	(−0.12, 1.00)
tetrachloroethylene	0.8	0.83	0.6	0.90	0.4	0.77	0.7	0.83
	(0.2, 25.1)	(−0.32, 1.00)	(0.1, 6.4)	(−0.19, 1.00)	(0.2, 3.0)	(−0.12, 1.00)	(0.2, 6.5)	(−0.30, 1.00)
toluene	12.5	0.65	7.3	0.86	7.5	0.83	8.3	0.77
	(2.3, 43.4)	(−0.98, 0.98)	(1.8, 18.5)	(0.22, 0.99)	(3.3, 23.8)	(−0.25, 1.00)	(2.7, 26.9)	(−0.67, 1.00)
trichloroethylene	0.2	0.88	0.2	0.88	0.2	0.40	0.2	0.69
	(0.0, 2.5)	(0.23, 1.00)	(0.1, 2.5)	(−0.17, 1.00)	(0.1, 0.5)	(−0.64, 0.99)	(0.1, 1.0)	(−0.26, 1.00)
o-xylene	2.0	0.92	0.9	0.92	0.7	0.82	1.1	0.90
	(0.4, 12.5)	(−0.09, 1.00)	(0.3, 4.0)	(−0.17, 1.00)	(0.4, 16.0)	(−0.24, 1.00)	(0.4, 12.9)	(−0.20, 1.00)
m-/p-xylene	5.6	0.90	3.1	0.93	2.2	0.71	3.5	0.86
	(0.9, 40.1)	(−0.16, 1.00)	(0.6, 11.8)	(−0.40, 1.00)	(1.3, 58.3)	(−0.26, 1.00)	(1.0, 40.1)	(−0.30, 1.00)

[a] Median root mean squared error ($\mu g/m^3$); n for number of research subjects = 25 for Battle Creek, 22 for East St. Paul, and 24 for Phillips. [b] Median correlation coefficient; n for number of research subjects varies from 20 to 23 for Battle Creek, $n = 22$ for East St. Paul, and n varies from 21 to 22 for Phillips. [c] 10th and 90th percentiles.

ethyl benzene, methylene chloride, styrene, toluene, trichloroethylene, tetrachloroethylene, o-xylene, and m-/p-xylene).

Measuring P exposures is the only way to determine unequivocally the magnitude, duration, and frequency of actual exposures experienced by people as they move through a variety of indoor and outdoor locations during their normal daily activities (1, 2). Consequently, P exposure is the de facto "gold standard" for assessment of individual and population exposures to VOCs. But because it is usually impractical and prohibitively expensive to measure P VOC exposures for everyone (or even a large sample of the population of interest), there is a continuing need to develop and validate practical and cost-effective surrogate estimators that are suitably accurate and precise. In this paper, we have examined the performance of three candidate estimators of P exposure: outdoor community concentration, indoor residential concentration, and a simple time-weighted model.

The evidence indicates that, consistent with previous studies in urban areas (10−15), P exposures tended to be higher than measured indoor concentrations, which tended to be higher than measured outdoor concentrations. The data for P and I concentrations also show that within-person variability for the 14 VOCs measured in this study typically spanned 1 or more orders of magnitude, while between-person variability usually spanned 2 or more orders of magnitude. These findings suggest that a substantial number of people and a substantial number of P and I measurements for each person may be necessary to adequately characterize VOC exposures for a particular population.

One of the novel aspects of this study was the use of MSE as well as correlation measures to assess the performance of multiple estimators (O, I, simple time-weighted model) for P exposure. To appreciate the value of MSE as a comparison metric at the subject-specific level, it is important to recognize the inherent limitations of R, a more traditional means of comparing exposure estimators.

To examine and compare estimators of P exposures, we assumed a set of measured P concentrations from a group of m subjects represented their actual exposures to an individual VOC. The vector of these observations is denoted as

$$\mathbf{P} = [p_{11} \cdots p_{1n_1}, p_{21} \cdots p_{2n_2}, ..., p_{m1} \cdots p_{mn_m}]$$

where p_{ij} gives the observed P exposure for the ith subject on the jth occasion. A candidate estimator of $\mathbf{P}$, denoted as $\hat{\mathbf{P}}_*$, is a vector of the same structure as $\mathbf{P}$ but with some function of the data at element $\hat{p}_{*ij}$. Metrics for assessing the ability of $\hat{\mathbf{P}}_*$ to estimate $\mathbf{P}$ traditionally include the sample correlation coefficient, R (28), which may be estimated for each subject over time or jointly for all subjects, where R takes values within the interval $[−1, +1]$.

Although R is a common metric for analyzing associations between P exposures and O or I concentrations, it is only a measure of the linear association between $\hat{\mathbf{P}}_*$ and $\mathbf{P}$ (29). To the extent that $\hat{\mathbf{P}}_*$ is an unbiased and precise estimator of $\mathbf{P}$, R approaches $+1$. However, R also approaches $+1$ in many other cases. For example, if $\hat{p}_{*ij} = (1/2)p_{ij}$ for all j, then $R_i = +1$ despite the fact that $\hat{\mathbf{P}}_*$ underestimates $\mathbf{P}$ by a factor of 2.

Another problem with R is its dependence on the distribution of $\mathbf{P}$ values. Despite having the same MSE in estimating $\mathbf{P}$, it can be shown that an estimator $\hat{\mathbf{P}}_*$ will yield potentially very different values of R, depending on the distribution of values of $\mathbf{P}$ that are sampled or selected for the study. Thus it is not possible to compare an estimator from one study with one from a different study, unless the sets of $\mathbf{P}$ measurements are the same or at least have similar distributions. Absent these common features, therefore, we cannot answer the question of which estimator is better.

In the calculation of a separate, longitudinal correlation coefficient for each subject, this dependence on the $\mathbf{P}$ distribution adversely affects the usefulness of the resulting
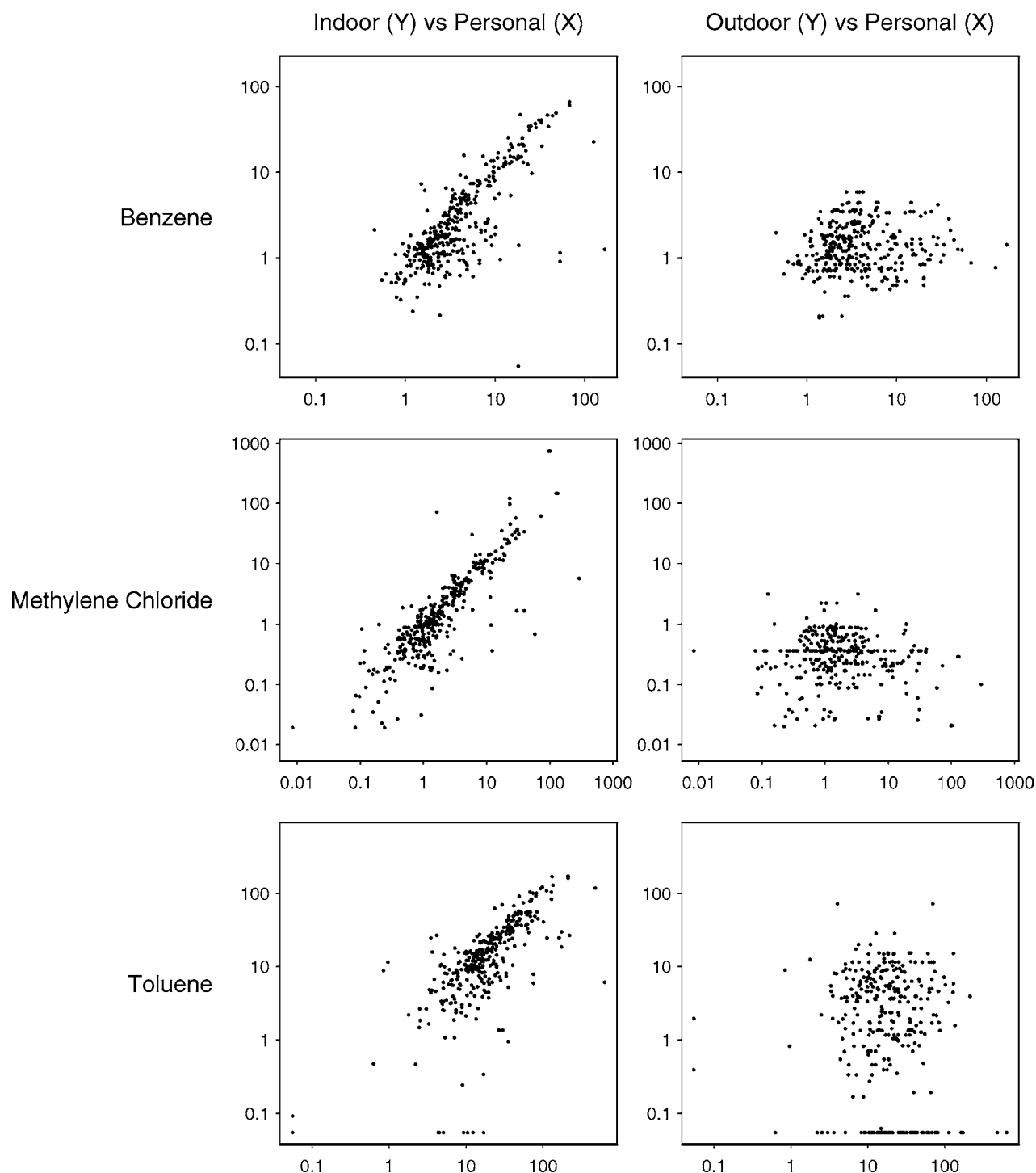
FIGURE 3. Plots of (a) indoor residential concentrations (μg/m^3) and (b) outdoor community concentrations vs personal exposures to benzene, methylene chloride, and toluene for participants in the study. As described in the text, nondetectable measurements (i.e., samples with no analytical response, or those with values $\leq$ 0 after blank subtraction) are represented with a value of half the analytical detection limit.

set of R_i values. The number and distribution of measured personal exposures will inevitably vary across subjects, but these factors should not bear on the assessment of the performance of, say, a monitor located at some central site. While the performance of this central site monitor may in fact be identical for all subjects, their own variation in personal exposure and compliance with the sampling effort can yield large differences in their longitudinal correlations. Finally, the use of longitudinal correlation as a comparison metric also means that subjects with only one measured VOC value cannot contribute an R_i. But in reality there is no

reason a single observation should not add to our understanding of the ability of $\hat{\mathbf{P}}_*$ to estimate $\mathbf{P}$.

For these reasons, we also assessed estimators of $\mathbf{P}$ in terms of MSE and its constituents: bias and variance. For the estimator of a specific element p_{ij} of $\mathbf{P}$, the bias of $\hat{p}_{*ij}$ for p_{ij} is the difference between the expected value of $\hat{p}_{*ij}$ and p_{ij}, i.e.:

$$\text{bias}[\hat{p}_{*ij}; p_{ij}] = E[\hat{p}_{*ij}] - p_{ij} \qquad (4)$$

where the $E[\ ]$ denotes the expectation operator. The variance

TABLE 4. Using Outdoor Community Concentrations To Predict Measured Personal Exposures: Estimated Bias and Variance (Presented as Standard Deviation in Parentheses) for the Lower, Middle, and Upper Tertiles of the Personal Exposure Distribution[a]

	personal exposure distribution				
chemical	lower tertile	1st tertile cutpoint	middle tertile	2nd tertile cutpoint	upper tertile
benzene	−0.3[b] (0.7)[c]	2.2	−1.5 (1.4)	4.7	−14.4 (21.9)
chloroform	−0.2 (0.2)	0.7	−1.0 (0.3)	1.7	−3.2 (1.9)
p-dichlorobenzene	0.0 (0.1)	0.2	−0.3 (0.2)	0.7	−7.8 (19.2)
ethyl benzene	−0.4 (0.5)	1.6	−1.5 (0.7)	3.5	−12.2 (19.3)
D-limonene	−4.9 (1.6)	7.7	−11.5 (2.7)	16.6	−47.3 (51.1)
methylene chloride	−0.1 (0.5)	1.0	−1.2 (0.6)	2.9	−17.1 (35.7)
α-pinene	−0.9 (0.4)	1.8	−2.6 (0.8)	4.2	−15.8 (20.6)
β-pinene	−0.2 (0.3)	0.8	−1.5 (0.5)	2.7	−10.9 (23.4)
styrene	−0.1 (0.1)	0.5	−0.5 (0.2)	1.0	−2.0 (2.8)
tetrachloroethylene	0.0 (0.3)	0.6	−0.6 (0.4)	1.5	−62.7 (360.4)
toluene	−1.9 (9.9)	12.1	−13.2 (6.4)	25.1	−57.8 (79.8)
trichloroethylene	0.0 (0.1)	0.2	−0.1 (0.2)	0.4	−2.4 (7.5)
o-xylene	−0.4 (0.6)	1.7	−1.6 (0.9)	3.8	−14.5 (20.4)
m-/p-xylene	−1.3 (1.7)	5.4	−5.1 (2.8)	12.4	−45.9 (66.2)

[a] All estimates and cutpoints in units of $\mu g/m^3$. [b] Bias estimated by the mean difference of predictor and personal exposure. [c] Variance estimated by the variance of the differences of predictor and personal exposure; with square root applied to present in terms of standard deviation (SD).

TABLE 5. Using Indoor Residential Concentrations To Predict Measured Personal Exposures: Estimated Bias and Variance (Presented as Standard Deviation in Parentheses) for the Lower, Middle, and Upper Tertiles of the Personal Exposure Distribution[a]

	personal exposure distribution				
chemical	lower tertile	1st tertile cutpoint	middle tertile	2nd tertile cutpoint	upper tertile
benzene	−0.2[b] (1.0)[c]	2.2	−0.6 (2.1)	4.7	−3.8 (20.8)
chloroform	0.4 (1.8)	0.7	−0.1 (0.6)	1.7	−0.5 (1.7)
p-dichlorobenzene	0.0 (0.2)	0.2	−0.2 (0.2)	0.7	−5.5 (19.4)
ethyl benzene	−0.2 (0.4)	1.6	−0.4 (1.1)	3.5	−4.7 (19.5)
D-limonene	−0.9 (2.0)	7.7	−1.6 (5.8)	16.6	−13.2 (50.1)
methylene chloride	0.0 (0.3)	1.0	1.1 (10.2)	2.9	3.9 (74.2)
α-pinene	0.0 (0.7)	1.8	0.0 (0.9)	4.2	0.8 (10.4)
β-pinene	0.3 (0.8)	0.8	−0.2 (0.9)	2.7	−2.1 (10.2)
styrene	0.1 (0.4)	0.5	−0.1 (0.3)	1.0	−0.8 (3.1)
tetrachloroethylene	0.0 (0.3)	0.6	−0.3 (0.4)	1.5	−55.8 (350.5)
toluene	0.2 (5.2)	12.1	−3.3 (8.2)	25.1	−19.9 (77.1)
trichloroethylene	0.0 (0.1)	0.2	0.1 (0.8)	0.4	−1.7 (7.6)
o-xylene	−0.2 (0.6)	1.7	−0.5 (1.2)	3.8	−5.3 (20.5)
m-/p-xylene	−0.6 (1.6)	5.4	−1.7 (3.9)	12.4	−17.0 (66.6)

[a] All estimates and cutpoints in units of $\mu g/m^3$. [b] Bias estimated by the mean difference of predictor and personal exposure. [c] Variance estimated by the variance of the differences of predictor and personal exposure; with square root applied to present in terms of standard deviation (SD).

TABLE 6. Using a Simple Time-Weighted Model To Predict Measured Personal Exposures: Estimated Bias and Variance (Presented as Standard Deviation in Parentheses) for the Lower, Middle, and Upper Tertiles of the Personal Exposure Distribution[a]

	personal exposure distribution				
chemical	lower tertile	1st tertile cutpoint	middle tertile	2nd tertile cutpoint	upper tertile
benzene	−0.2[b] (0.8)[c]	2.2	−0.8 (1.4)	4.7	−7.5 (21.0)
chloroform	0.2 (1.1)	0.7	−0.4 (0.4)	1.7	−1.2 (1.5)
p-dichlorobenzene	0.0 (0.1)	0.2	−0.2 (0.2)	0.7	−6.6 (19.4)
ethyl benzene	−0.3 (0.4)	1.6	−0.7 (0.8)	3.5	−6.9 (19.4)
D-limonene	−2.1 (1.8)	7.7	−4.4 (4.6)	16.6	−20.8 (48.6)
methylene chloride	0.0 (0.3)	1.0	0.5 (7.9)	2.9	−3.3 (48.2)
α-pinene	−0.3 (0.5)	1.8	−0.7 (0.7)	4.2	−4.8 (9.4)
β-pinene	0.1 (0.6)	0.8	−0.6 (0.7)	2.7	−5.6 (12.0)
styrene	0.0 (0.3)	0.5	−0.2 (0.2)	1.0	−1.2 (2.8)
tetrachloroethylene	0.0 (0.2)	0.6	−0.4 (0.3)	1.5	−60.9 (363.5)
toluene	0.0 (5.7)	12.1	−6.2 (5.9)	25.1	−30.6 (78.6)
trichloroethylene	0.0 (0.1)	0.2	0.0 (0.6)	0.4	−2.0 (7.7)
o-xylene	−0.3 (0.5)	1.7	−0.8 (0.9)	3.8	−8.1 (20.2)
m-/p-xylene	−0.8 (1.6)	5.4	−2.7 (2.7)	12.4	−26.0 (66.0)

[a] All estimates and cutpoints in units of $\mu g/m^3$. [b] Bias estimated by the mean difference of predictor and personal exposure. [c] Variance estimated by the variance of the differences of predictor and personal exposure; with square root applied to present in terms of standard deviation (SD).

of $\hat{p}_{*ij}$ does not depend functionally on p_{ij} and may be expressed as

$$\text{var}[\hat{p}_{*ij}] = E[(\hat{p}_{*ij} - E[\hat{p}_{*ij}])^2] \qquad (5)$$

The MSE is yet another linear operator comprising these constituents as

$$\text{MSE}[\hat{p}_{*ij}; p_{ij}] = (\text{bias}[\hat{p}_{*ij}; p_{ij}])^2 + \text{var}[\hat{p}_{*ij}] \qquad (6)$$

The bias and variance describe different characteristics of the estimator $\hat{p}_{*ij}$. Bias describes the extent to which $\hat{p}_{*ij}$ under- or overestimates p_{ij}. Variance conveys the precision of $\hat{p}_{*ij}$; the precision of a statistic is sometimes defined specifically as the inverse of its variance.

Based on MSE as well as R (Tables 2 and 3), I concentrations were a better estimator of P exposure than O concentrations for all 14 VOCs, although both consistently underestimated P exposure. There are several reasons for this. First, personal exposures tended to be higher than matched indoor residential concentrations, which tended to be higher than matched outdoor community concentrations. For example, median and 90th percentile values for benzene were 3.2 and 18.3 μg/m^3 in personal air, 1.9 and 15.3 μg/m^3 in indoor air, and 1.3 and 3.3 μg/m^3 in outdoor air (13). Second, most participants typically spent the majority of their time indoors at home (and relatively little outside). Results from the participants' 2-day time–activity logs show that, on average, participants spent 34 h (70.9%) indoors at home. The rest of the time was spent indoors at work or school (6 h or 12.6%), indoors in other locations (2.6 h or 5.5%), outside at home (1.7 h or 3.5%), outside at work or school (0.3 h or 0.6%), outside at other locations (1.1 h or 2.4%), and in transit (2.2 h or 4.5%). In addition, participants were in close proximity to a smoker for an average of only 0.5 h (0.9%) over a typical 2-day monitoring period. Third, the measured indoor concentrations may be an underestimate of what people were actually exposed to during their time inside at home. The monitors collected a 2-day integrated sample, but concentrations may have been highest when people were cooking and carrying on other routine activities. And fourth, it is possible that concentrations in other microenvironments through which participants moved during the 2-day monitoring period were relatively high as compared to measured I and O concentrations. Thus, although participants spent a relatively small proportion of their time indoors at work/school, indoors in other locations, outside at work/school, outside at other locations, and in transit, concentrations in these microenvironments appear to make a significant contribution to measured P exposure.

The bias and variance of all three estimators (indoor, outdoor, and time-weighted model) tended to increase in the upper third of the P exposure distribution. This means that common exposure estimators, such as measured indoor and outdoor concentrations and time-weighted models, tend to be less accurate and precise just where we need them most—for estimating exposures at the upper end of the exposure distribution. Future research should investigate whether these same patterns and relationships hold for (a) communities with higher outdoor levels of VOCs, (b) a more diverse sample of adults (race/ethnicity, socioeconomic status, occupation), and (c) vulnerable segments of the population (pregnant women and their fetuses, children, the elderly, the infirm).

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